AMENDED CLAIMS

[received by the International Bureau on 14 July 2004 (14.07.2004);
original claims 1-4, 11, 14-17 and 21 amended; remaining claims unchanged (5 pages)]

+ STATEMENT

AMENDED CLAIMS UNDER ARTICLE 19

1. A flameless distributed combustion heated, membrane, steam reforming reactor comprising:

a) a reforming chamber containing a reforming catalyst bed, said reforming chamber having an inlet for vaporizable hydrocarbon and steam, a flow path for hydrogen and by-product gases resulting from the reforming reactions taking place in said reforming chamber and an outlet for said by-product gases,

b) at least one flameless distributed combustion chamber in a heat transferring relationship with said reforming catalyst bed whereby a distributed, controlled heat flux is provided by said flameless distributed combustion chambers to said reforming catalyst bed, said flameless distributed combustion chamber comprising an inlet and a flow path for an oxidant, an outlet for combustion gas and further comprising a fuel conduit having an inlet for fuel and a plurality of fuel nozzles which provide fluid communication from within the fuel conduit to the flow path of said oxidant, said plurality of fuel nozzles being sized and spaced along the length of said fuel conduit so that no flame results when said fuel is mixed with said oxidant in said flameless distributed combustion chamber;

c) a preheater capable of preheating said oxidant to a temperature that when said fuel and said oxidant are mixed in said flameless distributed combustion chamber, the temperature of the resulting mixture of said oxidant and fuel exceeds the autoignition temperature of said mixture; and

d)at least two hydrogen-selective, hydrogen-permeable, membrane tubes in contact with said reforming catalyst bed, each of said membrane tube having an outlet whereby hydrogen formed in the reforming chamber permeates into said membrane tube and passes through said outlet.

2. A process for the production of hydrogen, comprising:

a) reacting steam with a vaporizable hydrocarbon at a temperature of from about 200°C to about 700°C and at a pressure of from about 1 bar to about 200 bar in a reaction zone containing a reforming catalyst to produce a mixture of primarily hydrogen and carbon dioxide, with a lesser amount of carbon monoxide;

b) providing heat to said reaction zone by employing at least one flameless distributed combustion chamber thereby driving said reaction; and
c) conducting said reaction in the vicinity of at least two hydrogen-permeable, hydrogen-selective membrane tubes, whereby hydrogen formed in said reaction zone permeates through said hydrogen selective membrane tubes and is separated from said carbon dioxide and carbon monoxide.

3. A membrane, steam reforming reactor comprising:
a) a reforming chamber containing a reforming catalyst bed, said reforming chamber having an inlet for vaporizable hydrocarbon and steam, a flow path for hydrogen and by-product gases resulting from the reforming reactions taking place in said reforming chamber and an outlet for said by-product gases,
b) at least one flameless distributed combustion chamber in a heat transferring relationship with said reforming catalyst bed,
and
c) at least two hydrogen-selective, hydrogen-permeable, membrane tubes in contact with said reforming catalyst bed, and at least one of the membrane tubes is also connected to a section containing a metal hydride precursor, and the hydrogen formed in the reforming chamber permeates through said membrane tube to said section containing the metal hydride precursor which reacts with the permeated hydrogen to form hydride.

4. A flameless distributed combustion heated, membrane, dehydrogenation reactor comprising:
a) a dehydrogenation chamber containing a catalyst bed, said dehydrogenation chamber having an inlet for vaporizable hydrocarbon, a flow path for hydrogen and product gases resulting from the dehydrogenation reactions taking place in said dehydrogenation chamber and an outlet for said product gases,
b) at least one flameless distributed combustion chamber in a heat transferring relationship with said catalyst bed whereby a distributed, controlled heat flux is provided by said flameless distributed combustion chamber to said catalyst bed, said flameless distributed combustion chamber comprising an inlet and a flow path for an oxidant, an outlet for combustion gas and further comprising a fuel conduit having

an inlet for fuel and a plurality of fuel nozzles which provide fluid communication from within the fuel conduit to the flow path of said oxidant, said plurality of fuel nozzles being sized and spaced along the length of said fuel conduit so that no flame results when said fuel is mixed with said oxidant in said flameless distributed combustion chamber;

c)a preheater capable of preheating said oxidant to a temperature that when said fuel and said oxidant are mixed in said flameless distributed combustion chamber, the temperature of the resulting mixture of said oxidant and fuel exceeds the autoignition temperature of said mixture; and

a) at least two hydrogen-selective, hydrogen-permeable, membrane tubes in contact with said catalyst bed, each of said membrane tube having an outlet whereby hydrogen formed in the dehydrogenation chamber permeates into said membrane tube and passes through said outlet.

5. A process for dehydrogenation of ethylbenzene, which process comprises the steps of feeding ethylbenzene into the reactor as described in Claim 4 to produce styrene and hydrogen.

6. A hydrogen fuel cell, wherein the hydrogen feed is made by a process as described in claims 2 or 5.

7. The reactor, process or fuel cell of claims 1, 2, 3, 4, or 5, wherein said catalyst bed is in contact with multiple hydrogen-selective, hydrogen-permeable membrane tubes.

8. The reactor, process or fuel cell of claims 1, 2, 3, 4, 5, 6, or 7, wherein said catalyst bed is in heat transferring contact with multiple flameless distributed combustion chambers.

9. The reactor, process or fuel cell of claims 1-8, wherein the vaporizable hydrocarbon and steam flow axially through said catalyst bed.

10. The reactor process or fuel cell of claims 1-8, wherein the vaporizable hydrocarbon and steam flow radially through said catalyst bed.

11. The reactor, process or fuel cell of claims 1-8, wherein a sweep gas is used to promote the diffusion of hydrogen through at least one of said membrane tubes, said sweep gas being selected from the group consisting of steam, carbon dioxide, nitrogen and condensable hydrocarbon and the vaporizable hydrocarbon is selected from the group consisting of natural gas, methane, ethyl benzene, methanol, ethane, ethanol, propane, butane, light hydrocarbons having 1-4 carbon atoms in each molecule, light petroleum fractions including naphtha, diesel, kerosene, jet fuel or gas oil, and hydrogen, carbon monoxide and mixtures thereof.

12. The reactor, process or fuel cell of claims 1-11 wherein the ratio of the surface area of said flameless distributed combustion chambers to the surface area of said membrane tubes is from about 0.1 to about 20.0, particularly from about 0.2 to about 5.0, more particularly from about 0.5 to about 5.0, and still more particularly from about 0.3 to about 3.0 and even more particularly from about 1.0 to about 3.0.

13. The reactor, process or fuel cell of claims 1-12 wherein said catalyst bed contains baffles in a form selected from the group consisting of (i) washers and disks, and (ii) truncated disks.

14. The reactor, process or fuel cell of claims 1-13, wherein the hydrogen-selective and at least one of the hydrogen-permeable membranes is made of a Pd-alloy layer supported on a porous metal, particularly a Pd-alloy layer deposited by electroless plating on porous metal with an in-situ oxide protection layer.

15. The reactor, process or fuel cell of claims 1-13, wherein at least one of the hydrogen-selective and hydrogen-permeable membranes is made of a Pd-alloy layer supported on a porous metal selected from the group consisting of (i) porous nickel-based alloy, (ii) porous Hastelloy®, and (iii) porous Inconel.

16. The reactor, process or fuel cell of claims 1-13, wherein at least one of the hydrogen-selective and hydrogen-permeable membranes has a ratio of length to diameter of less than about 500, gaps between the membrane tubes are from about ¼ inch (about

0.64 cm) to about 2 inches (about 5.08 cm), and gap between the membrane and FDC tubes is from about ¼ inch (about 0.64 cm) to about 2 inches (about 5.08 cm).

17. The reactor, process or fuel cell of claims 1-13, wherein at least one of the hydrogen-selective and hydrogen-permeable membranes has a ratio of length to diameter of less than about 250, gap between the membrane tubes is from about ½ inch (about 1.27 cm) to about 1 inch (about 2.54 cm), and gap between the membrane and FDC tubes is from about ½ inch (about 1.27 cm) to about 1 inch (about 2.54 cm).

18. The reactor, process or fuel cell of claims 1-17, wherein the FDC chamber having an external tubular dimension such that the length to diameter ratio is higher than 4.

19. The reactor, process or fuel cell of claims 1-17, wherein the FDC chamber having a tubular dimension such that the length to diameter ratio is higher than 10.

20. The reactor, or process of claims 1-3, wherein carbon dioxide produced from said steam reforming chamber has a pressure of from about 0.1 to about 20 MPa, particularly from about 1 to about 5 MPa based on an international standard.

21. The reactor or process of claims 1-3, wherein carbon dioxide produced from said steam reforming chamber has a concentration of from about 80% to about 99% molar dry basis.

22. The reactor, process, or fuel cell of claims 1-3, wherein carbon dioxide produced from said steam reforming chamber has a concentration of from about 90% to about 95% molar dry basis.

23. The reactor, process, or fuel cell of claims 1-3, wherein carbon dioxide produced from, said steam reforming chamber is used at least in part for enhanced recovery of oil in oil wells or enhanced recovery of methane in coal bed methane formations.